Response to second review

We would like to thank reviewer #1 for those helpful suggestions and comments which will help improving this manuscript.

Vernier et al. evaluated aerosol data collected on balloons launched into the Asian tropopause region during flight intensive flights (and one test flight). The authors generally have done a good job addressing many of the concerns raised by both reviewers, improving the paper. However, there are still some comments, as listed below, the authors should address before the paper should be accepted to ACP.

1) Page 2, Line 15 - 16: Please rephrase to "since NAT has already been observed in the tropical upper tropopsphere and lower stratosphere in other studies."

This is now corrected.

2) Page 7, Line 17: remove the m in prepared

Corrected.

3) Page 8, Line 6 - 7: Please rephrase to "two flights samples collected during the summer 2017 campaign (ZF2, 15th Aug. and ZF3, 21st Aug.), in comparison"

Corrected.

4) Pag 9, Line 21 - 23: Please rephrase to "The derived particulate depolarization ratio from CALIOP level 2v4.1 within the layer was 0.47+/-0.06 (Fig. S3) and was associated with an optical depth of 0.03+/-0.02, indicating the presence of a subvisible cirrus cloud."

Corrected.

5) Section 4.1. I am slightly confused by the reasoning here that HNO3 is transported into the UTLS instead of being locally produced. E.g., Bela et al. (2016) found nearly 90% scavenging efficiency for HNO3 in convection. Reconciliation of this high scavenging efficiency needs to be addressed.

Yes, however scavenging efficiency reported by Bela et al. (2016) was based on aircraft measurements over the continental United-States in the upper troposphere near 10-12 km while our measurements took place at higher altitudes in more polluted conditions where the transport of NOx is expected to be higher with a potential production of HNO3 in the Upper Troposphere and Lower Stratospheric region. We added a line about this. Section 4.1 has been modified accordingly.

6) Page 10, lines 5 - 6: It is unclear what "buffering process results in nitrate naturalization" means.

Line 5-6 has been removed since it was already discussed in line 25-26 p9.

7) Page 10, line 16 - 17: Please correct the capitalization of In after However

Corrected.

8) In all new text and throughout paper, please check the subscript of numbers and letters after chemical formulas (e.g., HNO2, NO2, NO3, NOx, etc.).

Corrected.

9) Addition of Sect. 5 is greatly appreciated; however, there are still some questions concerning it:
9a) As authors stated, K+ is signature for biomass burning. Why is no K+ observed?

We do not have a full explanation about this. Measurements in PyroCbs smoke plume are extremely rare and the chemistry not fully understood. While K+ is an element indicating the signature of biomass burning, we do not really know if it's still the case 6 months after a fire.

9b) NOx is normally emitted at high concentrations with biomass burning. Why is there no nitrite and nitrate?

We would propose the same argument than above.

9c) Also, it's surprising that SO4 is so high in this plume, when it has generally been observed that SO4 is a minor component of biomass burning aerosol. Why do the authors think SO4 is the major anion?

We believe that the BB plume could have been mixed with stratospheric sulfate which is a dominant component of stratospheric aerosol. In addition, PyroCbs plumes have also shown to contain the signature of tropospheric species and thus, we believe that SO2 as well as dust could have been transported in the UTLS.

10) Page 13, Line 1, please correct (1989 to (1989)

Corrected.

11) Page 13, Line 7: It is unclear what the authors mean that HNO2 undergoes rapid reduction. One of the challenges in measuring gas-phase HNO2, beyond wall loss as the authors note, is it's short lifetime due to photolysis, meaning it generally is at very low concentrations away from point sources. Please clarify.

We modified this sentence and added something along this line.

12) Page 13, line 15 - 16: It is unclear why the authors mention nitrification. Are they suggesting that nitrification in the soils produces enough HNO2 to be observed in the UTLS or that nitrification is occurring in the UTLS?

We argue that the presence of NH3 produced by agricultural activities combined with NOx can be potentially transported in the UTLS by convection and form nitrate.

13) Page 13, Line 24 - 29 and Page 14, Line 1 - 27: This should be in the methods section.

We decided to keep this section here since we believe that the GEOS-Chem simulation and its comparison with our measurements are part of the discussion and would thus read better where it is.

14) Page 15, Line 4 - 5: From the figure, it is unclear that BC is increasing. Instead, it appears BC has decreased in the region of the balloon launches. Please clarify.

Yes, it was a mistake which is now corrected.

15) Fig. 7, it would be beneficial to include a vertical bar for the flights

Now included

16) Page 15, Line 6: correct first

Sorry, we could not understand which correction is suggested here.

17) RC1.3 response. ISORROPIA has been evaluated extensively with in-situ nitrate observations, e.g., Guo et al., 2016, Guo et al., 2017, and Ibikunle et al., 2020, to name a few.

Thanks for pointing those references. They have been included in the manuscript.

References:

Bela et al., Wet scavenging of soluble gases in DC3 deep convective storms using WRF-Chem simulations and aircraft observations, JGR, 2016.

Guo et al., Fine particle pH and the partitioning of nitric acid during winter in the northeastern United States, JGR, 2016.

Guo et al., Fine particle pH and gas-particle phase partitioning of inorganic species in Pasadena, California, during the 2010 CalNex campaign, ACP, 2017.

Ibikunle et al., Fine particle pH and sensitivity to NH3 and HNO3 over summertime South Korea during KORUs-AQ, ACPD, 2020.